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FARMING PRACTICES AND CONCENTRATIONS OF FISSION PRODUCTS IN MILK

Warren G. Hansen, et al

Saint Louis County Health Department
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FARMING PRACTICES AND CONCENTRATIONS OF FISSION PRODUCTS IN MILK

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ABSTRACT

Studies conducted over a 3-year period have shown that levels of strontium-90 and iodine-131 in milk can be moderated by optimal fertilization of the land. In controlled experiments, milk from cows grazing on abundant, well-fertilized pastures contained 50 percent or less of these radionuclides than did milk from cows grazing on unfertilized pastures on the same farm. A mechanism of dilution of the fission-product contamination in the larger volume of faster growing fertilized forage was shown to account for the differences in the milk. The findings suggest one possible means of controlling fission-product contamination in milk, if such control measures should be necessary. The results indicate that the high levels of strontium-90 in the St. Louis raw milk samples may have been due more to marginal fertilization of the farms than to uneven deposition of fallout.

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INTRODUCTION

Iodine-131 and strontium-90 are fission products of paramount concern to human health--especially in the case of children who consume large quantities of milk, which is the chief source of both of these radionuclides in their diet. Although there is no evidence at present that prevailing levels of these radionuclides constitute a serious hazard to human health, it is prudent to maintain the intake at the lowest possible levels, in accordance with accepted public health principles. If atmospheric testing of nuclear devices should again occur, there may ultimately be a need to provide a means of reducing fission-product radioactivity in the human diet. In anticipation of such needs, several possible countermeasures in respect to milk have been considered, including ion exchange procedures, use of uncontaminated feeds, and diversion of milk supplies.

Although ion exchange columns to selectively remove these radioactive elements from milk are in process of development (1, 2), this method is envisioned only for emergency conditions. It has also been reported that milk from cows consuming only stored feed contained no detectable radioiodine while milk from cows grazing on comparable pastures during the same period contained from 50 to 270 pc per liter of iodine-131 (3). In dairying areas where stored feeds are available throughout the year, this finding could very readily be applied as a countermeasure. In other areas of the country, however, prevailing circumstances may prevent the use of this countermeasure at all times of the year. For example, in the dairying region of Missouri most dairy farmers have completely exhausted their stores of hay and silage by mid-May and must rely upon pastures for forage until the following fall. In any case, this method may not make an appreciable difference in the transfer of the long-lived strontium-90 from the environment, since it will be present in feeds in essentially the same concentration after a period of several

months of storage as it was at the time of harvesting. Diversion of milk supplies appears applicable only under limited conditions.

This report presents the results of an experimental inquiry into the possible relationships between certain farming practices and the levels of iodine-131 and strontium-90 in milk. Application of the experimental findings as a countermeasure can result in substantial reduction in levels of both iodine-131 and strontium-90 in milk without deleterious consequences to either the dairy industry or milk consumer. The specific farming practices that are of significance and the circumstances in which the experimental findings may be applied will be described.

PLAN OF STUDY

The work in the field was divided into two phases--survey and experimental. The survey phase, which was conducted from March 1960 to November 1961, was designed to isolate from the many variables involved in dairy farming the specific practices that might bear a relationship to levels of strontium-90 and iodine-131 in milk. The experimental phase, which was conducted during the grazing season (May to November) of 1962, was planned to verify the survey findings experimentally and to determine whether levels of iodine-131 and strontium-90 in milk could be moderated through the modification of farming practices.

Design of Surveys

In the survey phase of this study, each of the 66 farms making up the 1,000-cow milkshed that contributed to the St. Louis raw milk sample--one of nine in the milk network monitored by the U.S. Public Health Service (4, 5)--was visited on five occasions: March, August, and November, 1960; March 1961; and June 1961. This milkshed was especially appropriate for study because the St. Louis raw milk sample had been consistently higher with respect to strontium-90 than the other areas in the same monitoring program.

At each of these farms, information was gathered pertaining to breed, number, and nutritional condition of cows; average milk production; types and quantities of feeds; types and qualities of pasture and other feed crops; types, amounts, and frequency of

fertilization of the land; land and crop management programs; and depth of soil. Samples of milk, feeds, and water were taken from each farm at each survey for radiochemical analysis and correlation with survey findings.

A set of criteria for grading the fertilization and land management practices observed on these farms, which evolved during the course of study, is presented in table 1. Variables that did not enter into the results are omitted. Although objective criteria were followed, some subjective judgments were necessary in evaluating these practices. The possibility of bias entering into such judgments was minimized in two ways. All final gradings were based on the combined opinions of three observers, each of whom studied the farms independently. In addition, two blind surveys were conducted in which the two groups of farms were graded and the milk sampled by the cooperating creamery company. The samples were forwarded to the laboratory for analysis but the grading of farm practices was withheld until the analytical tests were complete. Then the farming practice ratings and the strontium-90 concentrations in milk were compared.

The survey phase was concluded during the fall of 1961 when 2 farms selected from the 66 were examined to determine what influence farming practices had on the degree of fission-product contamination in milk. This survey occurred during a period when the atmosphere contained fallout from the Soviet nuclear tests, and the milk contained measurable quantities of iodine-131. One of the farms, No. 164, is graded Good to Very Good by the criteria set forth in table 1. The pasture crops were in very good condition in late fall, furnishing ample forage for the grazing cows. The other, No. 161, is graded Very Poor by most of the criteria. The wild grasses and fescue in the permanent pastures as well as the wheat in the auxiliary pasture were exceedingly short as a result of the infertility of the land. (Fertilization practices on this land are described in the following section, Design of Experiment.) Samples of milk taken daily from these farms over a 3-week period were examined for iodine-131 and strontium-90 and the results compared. Samples of pasture forage were also taken daily. Most of these were forwarded to the Robert A. Taft Sanitary Engineering Center to be examined for iodine-131; four sets of the samples (taken on October 25 and 26, and November 2 and 6, 1961) were retained by the St. Louis County Health Department laboratory to be examined for strontium-90.

Table 1. Evaluation of Farming Practices According to Criteria of Study

	Classification				
	Very Poor	Poor	Fair	Good	Very Good
A. Farming practices (amounts of fertilizer in lb/acre)					
Pasture or hay	Rarely or never	Minimally every 3 to 5 yrs.	200 lb. or more of 12-12-12 when planted; top dressed at least every 2 yrs.	According to soil analysis when planted: Top dressed every 2 yrs.	According to soil analysis when planted: Top dressed annually
Silage crops corn or sorghum	150 lb. or less of 12-12-12 when planted	150 - 200 lb. of 12-12-12 when planted	Over 200 lb. of 12-12-12 when planted.	According to soil analysis when planted: Not side dressed.	According to soil analysis when planted: Side dressed during growing season
B. Land and Crop management practices					
Type of pasture	Wild grasses or fescue only		Permanent pasture of appropriate mixture of domestic grasses and legumes, in absence of auxiliary pasture	Similar, but in conjunction with auxiliary pasture: Auxiliary pasture in spring & fall of wheat or other appropriate crop; when in conjunction with legume-grass permanent pasture	Similar, but in conjunction with auxiliary pasture: Auxiliary pasture in summer of sudan grass or other drought-resistant crop; when in conjunction with legume-grass permanent pasture & auxiliary spring & fall pastures
Type of hay	Corn or sorghum in loose shocks	Wheat, oats, or rye; or timothy or other grass, no legumes	Domestic grasses and legume mixture. Fair or good, depending on how cured	Alfalfa or clover; good or very good depending on how cured	

Design of Experiment

From the 66 farms, 2 were selected for the experimental phase of this study: Farm No. 161 and farm No. 129 (table 2).

On farm No. 161, which had been closely surveyed during the fall of 1961, 61 acres of pasture were divided into control and experimental portions. The object of study on this farm was to determine whether the strontium-90 and iodine-131 concentrations of the milk could be reduced through the modifications in farming practices described below.

Thirty-five acres were selected as control pasture. This land, which had either been fertilized or had received minimal dressings of fertilizer several years previously, was in keeping with the farmer's usual management practices. The results of analysis of soil samples submitted to the local Agricultural Extension Agent indicated depletion of nitrogen, phosphorous, lime, and (on all but a 6-acre section) potash; organic content was low. Forage on this land consisted of a mixture of wild grasses, fescue, hop clover, and annual lespedeza. Even though the area experienced a severe drought during midsummer, the control pasture had at all times sufficient forage to meet the grazing needs of the 12 cows assigned to it.

The remaining 26 acres were designated experimental pasture and were isolated from the rest of the farm by lanes and fences of barbed wire. Samples of soil taken from various portions of this section were submitted to the local Agricultural Extension Agent for analysis to determine fertilizer requirements. Lime and fertilizers were applied to this land in accordance with the recommendations indicated by the soil tests, and included the following materials per acre: 3 tons of crushed limestone and 600 pounds of slaked lime, 400 pounds of "triple superphosphate," 100 pounds of potash, and 210 pounds of ammonium nitrate. A 7-acre section of this experimental pasture received an additional 140 pounds of ammonium nitrate per acre to meet the requirements of the sudan grass planned for that area. When crops were planted, an additional "booster" of 150 pounds per acre of 12-12-12 fertilizer was drilled in with the seeds. Four crops were planted in the experimental pasture and separated by electric fences. These included an auxiliary spring pasture of wheat which was used through May 31, 1962, an auxiliary summer pasture of sudan grass which was used from June 1 through September 30, 1962, an auxiliary

Table 2. Summary of Experimental Design

Farm No.	Group of cows	Time period	Grazing procedure
161	A	May to mid-June	Experimental pasture, fertilized as recommended by County Agent; crops planned to insure abundant forage at all times
	B	May to mid-June	Unfertilized control pasture; soil depleted of plant nutrients
	A	Mid-June to late October	Control pasture
	B	Mid-June to late October	Experimental pasture
	A	Late October to mid-November	Experimental pasture
	B	Late October to mid-November	Control pasture
129	C	May to mid-November	Well-fertilized control pasture
	D	May to mid-November	Unfertilized, virgin pasture
5 control farms	With high radionuclide content in milk	May to mid-November	Poor to Very Poor pastures
5 control farms	With low radionuclide content in milk	May to mid-November	Good to Very Good pastures

fall pasture of wheat which was used from October 1 through November 15, 1962, and a new permanent pasture consisting of orchard grass, timothy, red clover, ladino clover, and self-seeding lespedeza. The latter was used from June 15 through November 15, 1962. This schedule accommodated changes in status of crops throughout the season. The fast-growing sudan grass was clipped once early in July.

The 24 milk cows were divided into two groups designated A and B. After they were separated early in May 1962 they did not intermingle throughout the course of the experiment. At the outset, group A was placed on the experimental pasture and group B on the control pasture. After about 6 weeks the groups were switched so that group A was on the control pasture and group B on the experimental pasture. Then late in October 1962 the groups were switched a second time, restoring them to the original arrangement. This procedure provided an internal control, ruling out the possibility that results might be due to a chance segregation treatment of the land. Composite milk samples were taken from each group of cows two to four times each week for radiochemical examination. This variation in sampling schedule was dictated partly by changing weather and crop conditions, which could have influenced the results, and partly by the capacity of the laboratory to accommodate samples for gamma spectroscopy.

All cows received a supplement each day of 16 pounds of commercially prepared ground dairy feed. Cows grazing the experimental pasture received a mixture containing either 12 or 18 percent protein and 49 percent carbohydrate. Cows grazing the control pasture received "Horse and Mule Feed," a mixture of ground grains containing 9 percent protein and 65 percent carbohydrate. The nutritional quality of the latter mixture approximated that of various mixtures observed on the farms graded Poor and Very Poor during the surveys. Feeding of supplements to individual cows, based on milk production, was not attempted.

The other farm selected for experimentation, No. 129, is graded as Very Good using the criteria of table 1. The object of study on this farm was to determine whether fission-product concentrations in milk could be increased by reversal of the experimental design utilized on farm No. 161. This study was useful as a confirmatory experience.

On farm No. 129, cows normally accustomed to abundant, well-fertilized pasture were grazed on unfertilized land. A group of 12

animals, representative of the entire herd, was placed on a 15-acre section of unfertilized, virgin grassland. Soil analysis of this section indicated deficiency in nitrogen, potash, and phosphorous; but adequacy in lime, and high content of organic matter. The remaining 20 milk cows were grazed on various pastures, all of which had received adequate amounts of fertilizer within a year, in accordance with the usual practices of the farmer.

On both farms, production of milk was recorded the same day each week on an individual cow basis. Two series of pasture samples were taken from both farms for radiochemical analysis. The first series consisted of three sets of samples taken at approximately 2-week intervals during June and July. These samples were collected as representative composites of an entire pasture by pooling many small portions of forage gathered from many scattered locations over the entire area. The second series of three sets of samples were taken at approximately 2-week intervals during September and October. These were taken by measuring out a single area of exactly 9 square feet on each pasture and cutting all of the edible forage as close to the ground as possible.

To serve as controls, 10 farms which had not been manipulated were selected. Of these, five had Poor or Very Poor programs of fertilization and land management and five had Good or Very Good programs. Composite samples of milk were taken from each of these farms for radiochemical examination at intervals of 3 weeks to a month and the results compared with those from the experimental farms.

Analytical Methods

Radiochemical analyses of all samples of milk from the first four surveys were performed at the Robert A. Taft Sanitary Engineering Center. All subsequent milk samples and all feed samples were analyzed at the St. Louis County Health Department. Strontium-90 was determined by chemical separation of radiostrontium and strontium carrier followed by extraction of the daughter, yttrium-90, counting in a low background anticoincidence beta counter, and determination of carrier yield by flame photometry (6). All strontium-90 assays were performed in duplicate. Iodine-131 was determined by gamma spectroscopy using 3.5-liter samples in Marinelli beakers placed on a 4- by 4-inch NaI (Tl) crystal contained within a steel shield 6 inches thick and coupled

to a 256-channel pulse height analyzer (7). Calcium was determined by oxalate precipitation and permanganate titration (8). Details of all analytical procedures are presented in appendixes I-IV.

RESULTS

The findings of these studies are presented in three sections. The principal observations from the surveys which provided the basis for the experimental work are presented first. These are followed by the results of the experimental procedures used on the two farms. Finally, certain observations made during the surveys are examined and interpreted in the light of the experimental findings.

Survey Findings

The surveys revealed that levels of activity of strontium-90 in milk from the 66 farms varied widely. On the first survey, the sample of milk with the lowest strontium-90 content contained only 6 pc per liter while the sample with the highest level contained 44 pc per liter. Similar variation was observed on each of the subsequent surveys. In a number of cases, farms with such contrasting levels of the radionuclide in the milk were located within a mile or two of each other. Not only was there wide variation between farms at any given survey, but also in a large number of cases the relative level of strontium-90 in milk from any given farm would vary from one survey period to the next. Because of these fluctuations from one survey period to the next, which characterized more than half of the 66 farms, consistent correlations by means of accepted statistical procedures between concentrations of strontium-90 in milk and the farming variables listed under Design of Surveys could not be obtained. Therefore, other methods of interpreting the data were explored. Using the extreme values obtained from all surveys, three categories--high, intermediate, and low--were established. The levels of strontium-90 in milk from 17 of the 66 farms remained in the same category each time the samples were assayed and are listed in table 3. An analysis of variance (9) showed that the differences, not only between the three consistent groups but also between the consistent groups and the more widely fluctuating groups, were highly significant at the level $p < 0.0005$. These 17 farms were examined

Table 3. Levels of Strontium-90 in Milk from Three Groups of Farms
(Concentrations in pc/1)

Farm Number	Survey Period: March 1960	August 1960	November 1960	March 1961	June 1961
Group I Consistently "High" Farms					
128	29.1	24.3	28.9	14.8	43.1
205	44.1	23.8	15.2	19.0	21.4
280	35.5	31.5	17.1	14.9	29.2
161	35.4	24.9	22.5	16.1	20.0
125	32.4	22.0	27.5	20.8	25.5
132	37.9	29.0	22.3	18.9	24.8
Average	35.8	25.9	22.2	17.4	27.3
Group II Consistently "Intermediate" Farms					
369	20.2	10.4	14.2	9.7	17.3
991	22.6	13.5	11.0	9.5	10.4
998	20.1	19.0	16.2	11.3	9.1
167	22.0	16.6	10.7	8.4	15.9
380	18.7	18.0	14.3	12.0	
109	24.2	15.5	11.9	12.3	12.4
Average	21.3	15.5	13.0	10.6	13.0
Group III Consistently "Low" Farms					
129	10.6	8.3	7.9	5.0	7.2
599	11.0	7.3	8.4	8.5	13.1
900	15.3	7.3	5.7		5.3
164	12.0	14.8	8.7	8.1	6.1
410	9.0	11.6	10.5	8.6	8.7
Average	11.6	9.9	8.2	7.6	8.1

for variables which might be related to each of the three categories--high, intermediate, and low. The remaining farms were culled from consideration until later in the study when a probable explanation for the fluctuations became evident. Study of these 17 farms suggested an inverse relationship between the quality of fertilization, along with management of the land, and the concentrations of strontium-90 in the milk. Levels of strontium-90 in milk, averaged over the five surveys, are compared (fig. 1) with

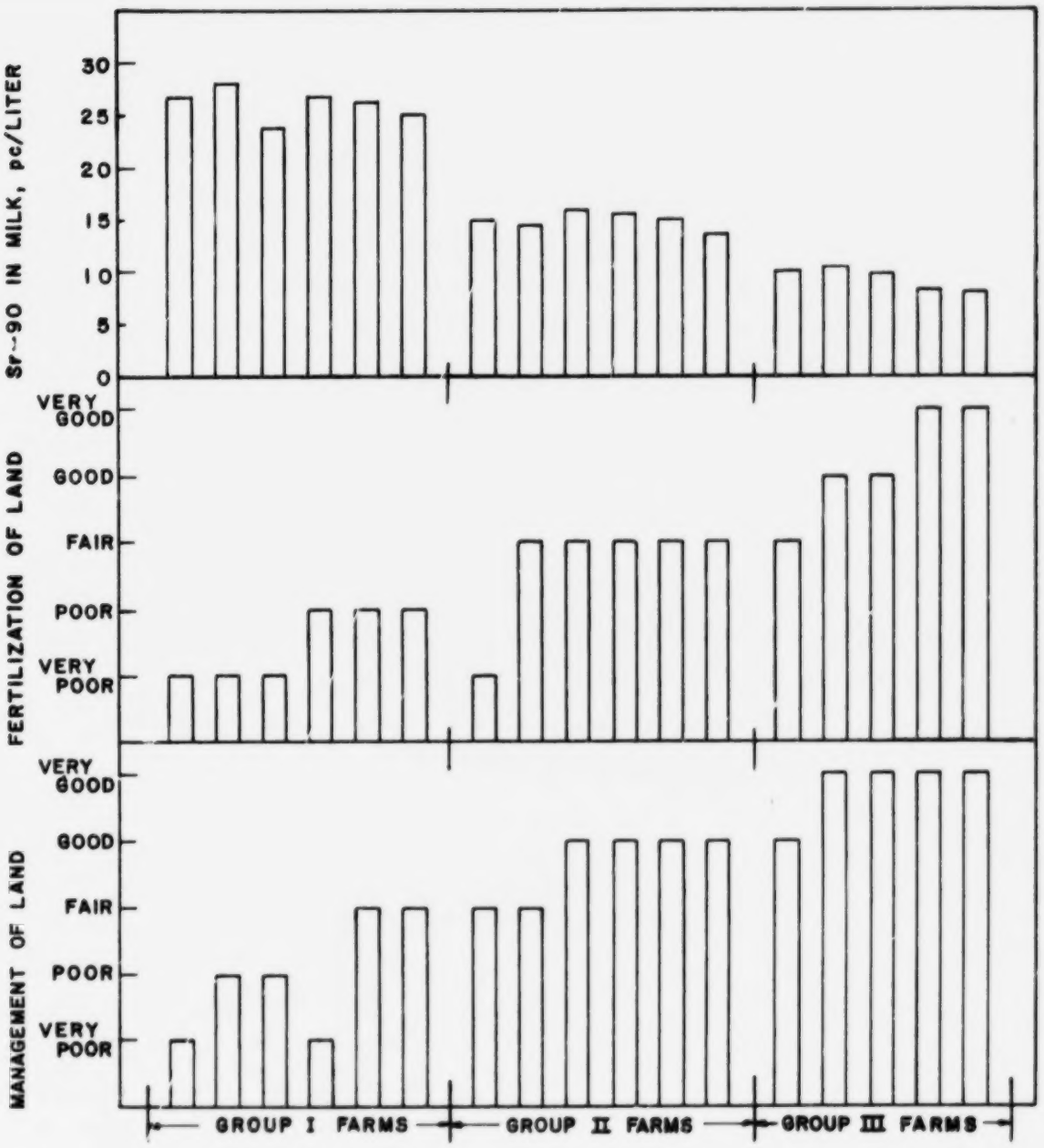

Figure 1. Comparison of management of the land (lower case) and fertilization of the land (middle case) with average levels of strontium-90 in milk (upper case) on three groups of farms

the gradings of fertilization and land management practices observed on these farm groups (according to the criteria established in table 1).

The inverse relationship between quality of fertilization of the soil and land management practices and strontium-90 content in the milk, which is clearly apparent in figure 1, was verified by the two blind surveys in which grading of fertilization and land management was furnished by the cooperating creamery company on the basis of their own knowledge and experience. The results

of the blind surveys are presented in table 4. Of the 12 farms classified as Fair or better, all but 2 had strontium-90 concentrations in milk of less than 12.0 pc per liter. Of the 11 farms classified as Inadequate in respect to management, all but 2 had strontium-90 concentrations in milk of 12.1 pc per liter or greater.

Analysis of feeds provided insight into the nature of the relationship between the quality of land management and fertilization and the levels of strontium-90 in milk. Hay, pasturage, and silage contributed most of the strontium-90 that appeared in the milk, while ground grain mixtures fed during milking always had relatively low concentrations of the radionuclide. This, of course, had a diluting effect on the total daily intake of radionuclides in feed. A quantitative evaluation of this diluting effect is prevented by the unreliability of information pertaining to proportions of different feeds. In all cases, levels of strontium-90 in the drinking water were too low (0-2 pc per liter) to contribute significantly to levels in the milk. In the majority of cases, concentrations of strontium-90 were greater in hay and pasture than in silage.

Table 4. Strontium-90 in Milk Related to Farm Management Gradings on Two Blind Surveys
(Concentrations in pc/1)

March 1961			June 1961		
Farm No.	Grading	Strontium-90	Farm No.	Grading	Strontium-90
99	A	1.8	211	A	6.6
106	A	5.4	134	A	12.8
122	A	8.8	72	B	10.5
100	B	11.3	53	B	11.6
66	B	11.9	266	C	8.9
218	C	8.9	215	D	9.4
346	C	19.2	150	D	12.1
198	D	9.3	16	D	12.5
402	D	14.6	213	D	12.8
41	D	15.9	165	D	21.9
195	D	17.0			
95	D	18.0			
10	D	24.9			

A - Excellent
B - Good
C - Fair
D - Inadequate

In figure 2, the average concentrations of strontium-90 in roughages (hay, silage, and pasturage) from the six "high" farms of group I are compared with the average concentrations in roughages from the five "low" farms of group III (table 3), and with average levels in the milk from these groups of farms at each of the survey periods. The relationship between the concentrations of the radionuclide in these feeds and the levels in the milk at each survey period is readily seen. It may be inferred from the data that the

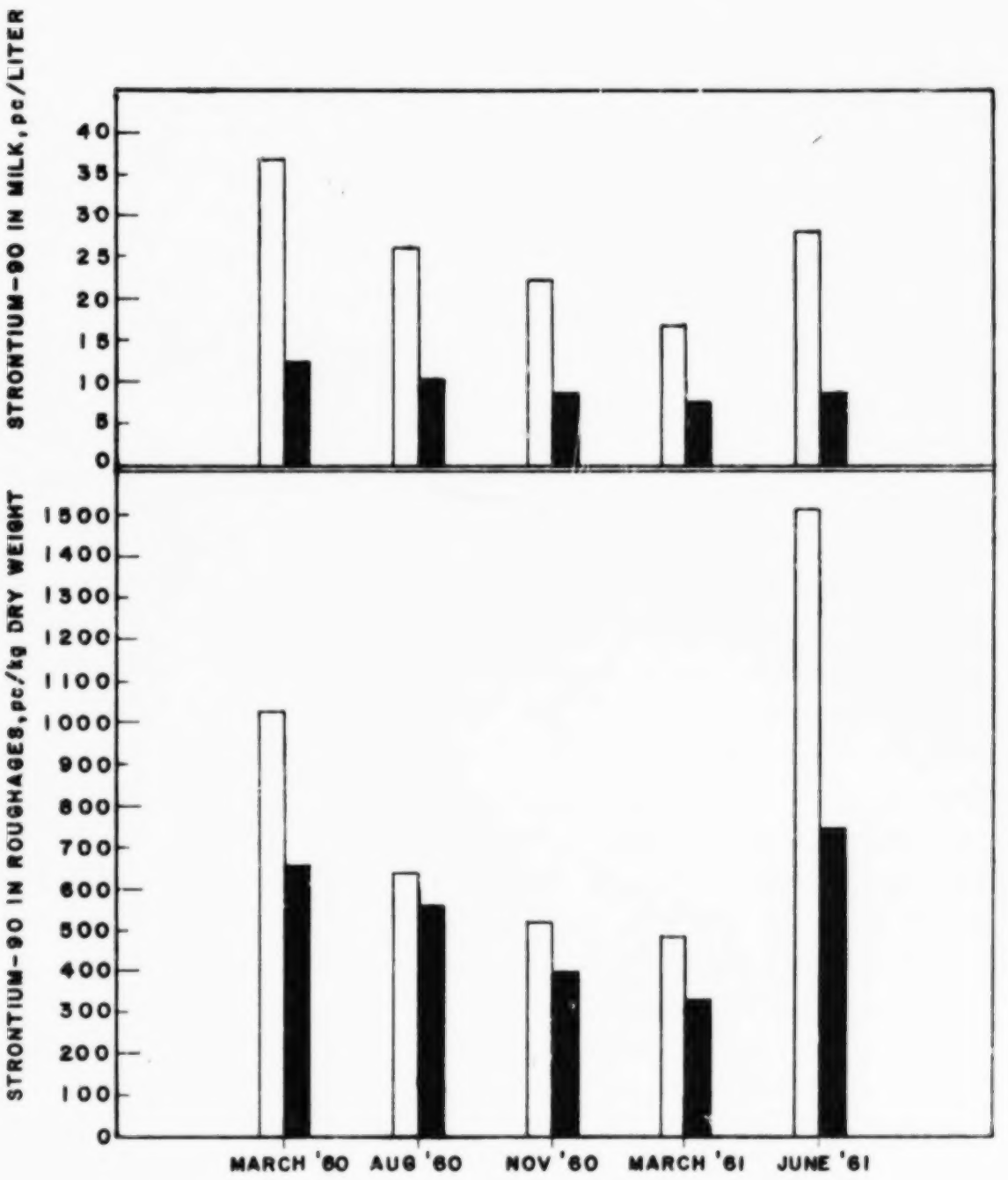

Figure 2. Strontium-90 in roughages (lower case) and strontium-90 in milk (upper case) from group I farms (open bars) and group III farms (solid bars)

six "high" farms of group I, which were graded Poor or Very Poor with respect to fertilization and land management, tended to produce forage containing higher concentrations of strontium-90 than the five "low" farms of group III graded Good or Very Good with respect to these variables. Figure 2 also shows that the differences in feeds are in turn reflected as differences in the milk.

The 3-week study during October-November 1961 of two adjacent farms provided a bridge between the survey phase of this study and the experimental phase that followed. The surveys had been conducted during the moratorium on nuclear testing. Since fresh deposition of fallout was declining during the survey period, it may be speculated that a significant fraction of the strontium-90 in feeds was due to root uptake from cumulative deposits in the soil (10). By contrast, it must be assumed that all of the iodine-131 and a significant fraction of the strontium-90 observed during the fall of 1961 resulted from fresh contamination on growing plants from the nuclear tests series conducted by the Soviet Union during that period. Somewhat surprisingly, since the two radioelements are metabolized in very different fashion, the levels of iodine-131 in milk from the two farms followed a pattern similar to that of levels of strontium-90 in milk (fig. 3) and were consistently lower in the case of farm No. 164 than in the case of farm No. 161. Both farms received essentially the same amounts of airborne beta activity during the study period. It is of interest to note that iodine-131 was not observed in the radioactive particulate matter collected by the high volume air samples. Farm No. 161 received about 13 percent less fresh fission-product deposition in rainfall (as estimated from the iodine-131 content of the rainwater) because of local variations in the intensity of storm activity.

Analysis of pasture revealed that concentrations of strontium-90 averaged nearly twofold higher in samples from farm No. 161 (which received a smaller deposition of fresh fallout in rain) than in samples from farm No. 164 (which received a larger deposition of fresh fallout in rain). These observations, shown in table 5, obviously account for the differences in strontium-90 levels in the milk from the two farms, and are in agreement with the earlier findings pertaining to feeds, depicted in figure 2. Unfortunately, the presence of large amounts of gamma-emitting radionuclides interfered with the radioassay of iodine-131 and prevented satisfactory analysis for that fission product in this group of pasture samples.

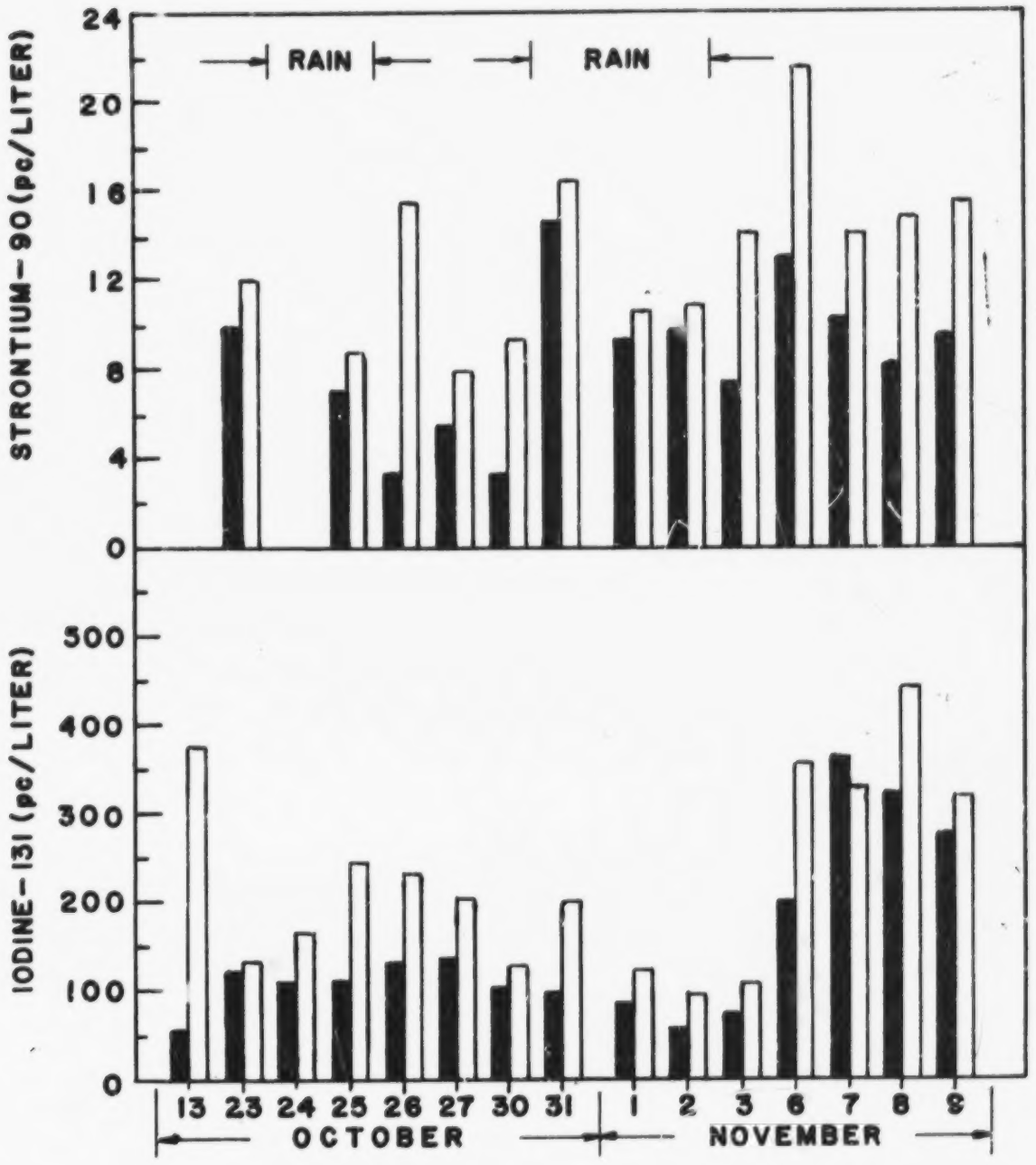

Figure 3. Daily levels of strontium-90 (upper case) and of iodine-131 (lower case) in milk from farm No. 164 (solid bars) and from farm No. 161 (open bars)

Table 5. Strontium-90 in Pasture on Two Closely Situated Farms
(Concentrations in pc/kg dry wt)

Date	Farm No. 164	Farm No. 161
October 25, 1961	612	659
October 26, 1961	561	956
November 2, 1961	620	1,990
November 6, 1961	1,126	1,910
Average	730	1,380

Recalling the description of the pastures on these two farms (Design of Surveys), it is possible to condense the findings of the earlier surveys with those of this 3-week study into a single hypothesis which may be stated as follows: Good management and fertilization of the land reduce the concentration of radioiodine and radiostrontium in milk through some mechanism involving growing feeds.

Experimental Results

Weekly averaged concentrations of iodine-131 and strontium-90 in milk from the two groups of cows on farm No. 161 are shown in figure 4. For reference, the location of each group of cows, the kind of experimental pasture utilized, and rainfall conditions are indicated. The 12 cows on the control pasture were permitted to roam over the entire 35-acre area. The 12 cows on experimental pastures grazed over 6 acres during the time they were on wheat pasture only, 7 acres during the time they were on sudan grass only, 20 acres during the time they were on sudan grass and new permanent pasture, and 19 acres during the time they were on the fall wheat pasture and new permanent pasture. As noted earlier (Design of Experiment), amounts of forage on the control pasture were at least adequate at all times for the grazing needs of the cows.

For ready comparison with results, the status of the pasture crops throughout the season is reviewed here. The control pasture during May was an abundant mixture of grasses and hop clover, averaging 6 inches high. During June, hop clover dominated. By the end of June, grasses were only 2 to 3 inches high. During the drought of July and August, lespedeza, which remained 3 to 6 inches high, provided most of the forage. With the return of rainfall in September, grasses rejuvenated and averaged 3 inches or more in height for the remainder of the experiment. In the case of the experimental pastures, the wheat was approximately 2 feet high early in May. During the last week in May, it headed out, then ripened, becoming unsuitable for grazing. Early in June, the sudan grass averaged 1 1/2 to 2 feet high. By the end of June, it was over 5 feet high, and heading out. It was clipped early in July and remained about 1 1/2 feet high during the drought. The new permanent pasture was about 1 foot high in mid-June when it was first used. By mid-July, it averaged 1 1/2 feet high. Through the drought,

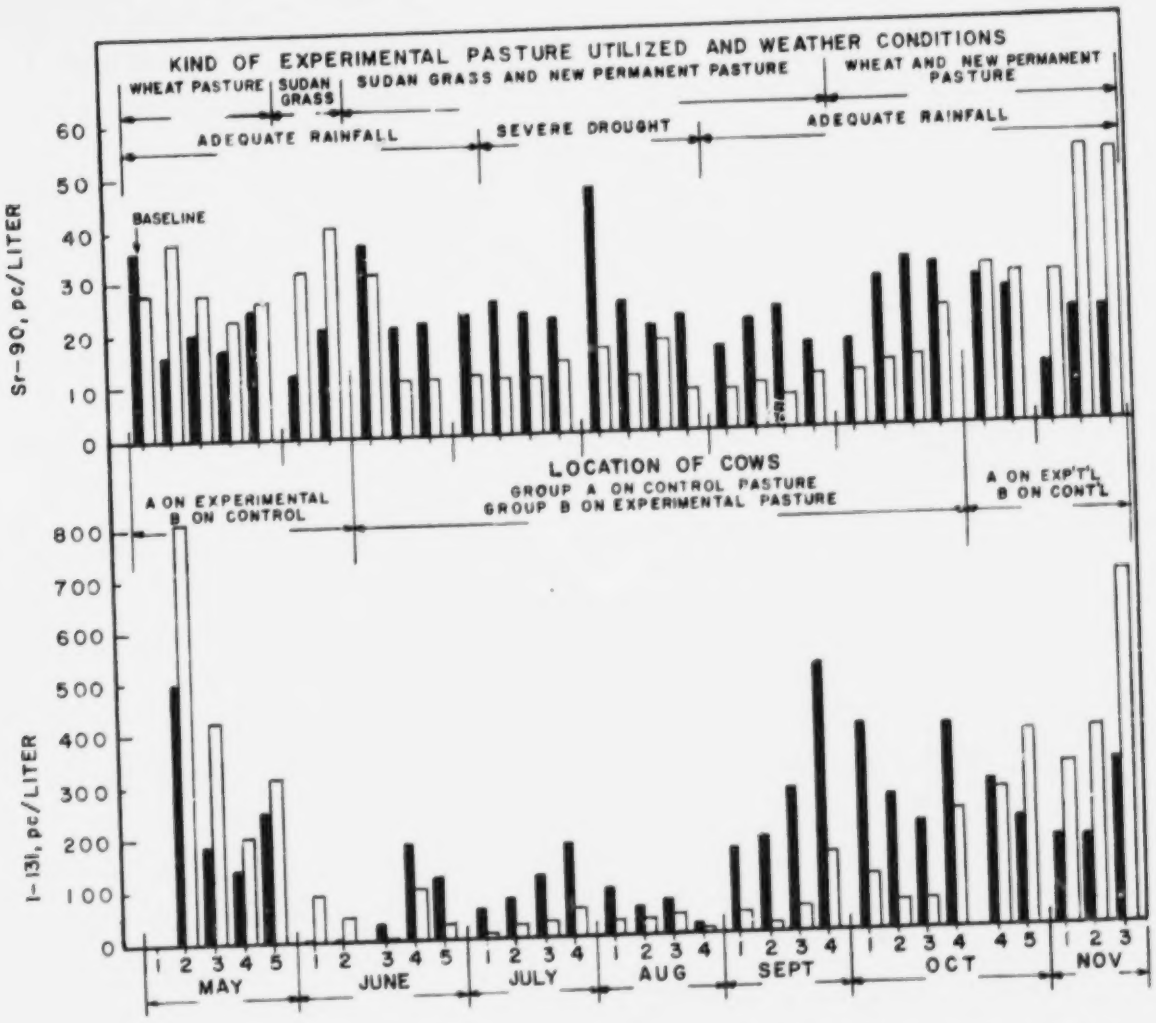

Figure 4. Weekly levels of iodine-131 and of strontium-90 in milk from group A cows (solid bars) and from group B cows (open bars) on farm 161.

forage on this section was 8 inches to 1 foot high. During August, an abundance of crabgrass (which is quite palatable to cows) appeared in some areas. With the return of rainfall, good growth maintained the forage crops at 1 1/2 to 2 feet high for the remainder of the season. The fall wheat maintained a range of 6 inches to 1 foot in height.

Figure 4 shows that levels of iodine-131 and strontium-90 were consistently substantially lower in milk from the cows which grazed the well-fertilized experimental pasture than in milk from the

cows grazing the unfertilized control pasture. It is of interest to note that the difference in the levels of strontium-90 and iodine-131 between group A and group B cows narrowed markedly during the fourth and fifth weeks of May, as the wheat pasture ripened and became unsuitable for grazing; then widened when the group A cows moved to the luxuriant pasture of sudan grass. The average level of iodine-131 (corrected for 4 days decay, which is the approximate time lag from milking parlor to metropolitan consumer) in milk from the cows grazing the experimental pastures was 69 pc per liter, less than half the average level of 163 pc per liter in milk from the cows grazing the control pasture. Similarly, the average level of strontium-90 in milk from the cows on the experimental pasture was 13.7 pc per liter, or approximately half the average level of 28.2 pc per liter in milk from the cows on the control pasture. It is important to note that all of the iodine-131 observed in these studies resulted from fresh deposition of radioactive debris from the nuclear tests conducted in 1962. (The first increments appeared in this area on May 8, 1962.) In environmental samples from this farm prior to the outset of the experiment, residual iodine-131 from the Soviet tests of the fall of 1961 had decayed to levels below detectability.

In arriving at an explanation for these results, several possibilities were ruled out or rendered highly unlikely. The procedure of switching the groups of cows, which served as an internal control, ruled out the possibility that the results might be due to a chance segregation of cows with unique metabolic capabilities. Although there was considerable fluctuation in levels of these fission products in milk of both groups of cows from time to time (fig. 4)--undoubtedly a function of rates of fresh deposition of fallout and availability to the cow--the relative levels in milk from the two groups of cows were not influenced by seasonal changes in weather. Although both groups of cows responded to the luxuriant forage of the fertilized experimental pasture by an increase in milk production, dilution of the radionuclides into a larger volume of milk was ruled out as an explanation of the findings. During the periods of July and August, daily average milk production by both groups of cows was essentially the same (table 6), while concentrations of iodine-131 in milk of the group A cows were approximately threefold higher than those in the group B cows, and the strontium-90 content of milk from the group A cows was about twice that in milk of the group B cows. Since the experimental and control

Table 6. Milk Production of Farm No. 161 Cows on Experimental and Control Pastures
(Averaged over 2-week periods)

Date	Group A (lb/cow/day)	Group B (lb/cow/day)
Before Segregation onto Experimental & Control Pastures		
May 2, 1962	22.2	18.9
Before the Groups Were Switched		
May 10, 1962	24.8	19.8
May 23, 1962	28.9	20.1
Jun. 7, 1962	30.7	21.5
After the Groups Were Switched		
Jun. 21, 1962	30.8	25.1
Jul. 4, 1962	29.8	26.6
Jul. 19, 1962	31.3	29.5
Aug. 2, 1962	28.1	28.1
Aug. 16, 1962	26.0	26.3
Aug. 29, 1962	22.9	23.0
Sep. 13, 1962	21.1	24.3
Oct. 24, 1962	21.1	23.5
After the Groups Were Switched Back		
Nov. 8, 1962	24.7	19.9
Nov. 15, 1962	25.3	19.6

pastures were separated only by fences and a thin strip of woods, it seems highly unlikely that the differences could be due to local climatic variations. The possibility that the cows on the experimental pasture might possess a superior ability to discriminate against strontium-90 was considered and ruled out insofar as these findings are concerned. The Observed Ratio as defined by Comar (12) was determined for both groups of cows and found to be the same. These results, shown in table 7, represent averages of all pasture samples and all milk samples taken from farm No. 161 during the study period. A diluting effect due to different intakes of ground feed was not a factor in explaining the differences in radionuclide levels in milk from the two groups of cows. As noted earlier (Design of Experiment) each group received the

Table 7. Strontium-Calcium Observed Ratios: Pasture to Milk on Farm No. 1[illegible]

	Cows on fertilized pasture	Cows on unfertilized pasture
Av. Sr^{90} in milk, pc/l	13.7	28.2
Av. Ca in milk, g/l	1.34	1.36
Strontium units in milk, Sr/Ca[1]	10.2	20.7
Av. Sr^{90} in pasture, pc/kg dry wt	1,020.	1,970.
Av. Ca in pasture, g/kg dry wt	4.95	4.88
Strontium units in pasture, Sr/Ca[1]	206.1	403.7
O R $\frac{\text{Sr/Ca in milk}}{\text{Sr/Ca in pasture}}$	0.050	0.051

[1] Ratio expressed as pc Sr^{90}/g Ca

same quantity of ground feed. Iodine-131 was not detected in either the commercial dairy ration fed to the cows on experimental pasture, or in the "Horse and Mule Feed" given to the cows on control pasture. Strontium-90 concentrations were not grossly different in these feeds, averaging 92 pc per kg dry weight in eight samples of dairy ration, and 73 pc per kg dry weight in eight samples of "Horse and Mule Feed."

These findings clearly demonstrate a causal relationship between fertilization and management of the land and levels of iodine-131 and strontium-90 in the milk. An indirect mechanism accounting for this relationship was found to be dilution of the radionuclides in the larger mass of fertilized forage. In nearly all cases, the concentrations of iodine-131 and strontium-90 in the fertilized forage from the experimental pastures were lower than in comparable samples of unfertilized forage from the control pastures. The average dry weight of pasture plants growing on a square yard of fertilized pasture was approximately twice that of the plants growing on a square yard of unfertilized pasture (table 8). However, the concentrations of iodine-131 and strontium-90 in the fertilized plants were about half the concentrations in the unfertilized plants.

Table 8. Average Weights and Radionuclide Content of Pasture Samples from Farm No. 161

	Fertilized experimental pastures	Unfertilized control pastures
Number of samples	8	6
Av. dry wt of forage g per sq yd	580	280
Av. I^{131}, pc per kg (dry wt)	794	1,590
Av. I^{131}, pc per sq yd of forage (dry wt)	460	450
Av. Sr^{90}, pc per kg (dry wt)	1,060	2,060
Av. Sr^{90}, pc per sq yd of forage (dry wt)	610	580

The same amounts of fission products per unit area were deposited and taken up by the plants on both experimental and control pastures.

It has not been determined whether the types of pasture plants in these studies take up the deposited fission products simply by adsorption of radioactive particulates onto the surfaces of leaves and stems, by foliar absorption (11), by root uptake (10), or by a combination of these. In any event, the more rapidly growing, well-fertilized plants distribute these fission products throughout a larger mass of plant substance, resulting in a lower concentration per unit of dry plant weight. When concentrations of iodine-131 and strontium-90 per unit of dry plant weight are expressed as content of these radionuclides in dry plant weight per square yard, the values are approximately the same (table 8). These observations agree with the earlier findings of the survey phase of this study and in large measure account for the lower levels of strontium-90 in feeds grown on poorly managed farms as compared with feeds grown on well-managed farms. If a cow grazing on unfertilized pasture consumes the same quantity of forage as a cow grazing on fertilized pasture, she will take in the fallout deposited over more than twice the area. The findings in the milk

may be accounted for entirely in terms of these observations. It is not necessary to evoke more subtle mechanisms for speculation. It is to be recognized, however, that other farming practices and conditions, such as deep plowing and high organic content of the soil, may also influence radionuclide levels in milk, independently of fertilization.

These observations are confirmed by the results of the reverse experiment on farm No. 129 and by the findings from the 10 control farms (table 2). The average levels of strontium-90 in milk from the two groups of cows on farm No. 161 are compared with the average levels of strontium-90 found in milk from the 10 non-manipulated control farms, and with the average levels of strontium-90 found in milk from farm No. 129 (table 9). The data from farm No. 129 clearly validate the conclusion that levels of strontium-90 in milk are influenced by quality of fertilization of the

Table 9. Comparison of Average Levels of Strontium-90 in Milk from Control Farms with Levels from Farms No. 129 and No. 161

(Concentrations in pc/l)

Farms with Good land management and fertilization programs			Farms with Poor land management and fertilization programs		
Farm No.	Sr^{90}	Number of observations	Farm No.	Sr^{90}	Number of observations
164	15.6	11	128	34.2	11
410	18.1	5	205	30.8	13
106	16.5	6	186	34.7	12
900	15.3	5	280	24.2	11
380	16.5	8	181	24.1	10
Average[1]	16.4			29.6	
Cows on Control Pasture			Cows on Experimental Pasture		
129	11.7	51		18.3	51
Cows on Experimental Pasture			Cows on Control Pasture		
161	13.9	49		28.2	49

[1] Results are averaged over the grazing season from May to November.

land, insofar as this governs the amount of forage produced per unit area. In the case of farm No. 129, the average concentration of strontium-90 in milk was nearly doubled when the experimental cows, ordinarily pastured on well-fertilized land, grazed on unfertilized land. In agreement with the findings on farm No. 161, the strontium-90 content of pasture forage from farm No. 129 was greater in the unfertilized than in the fertilized pastures (table 10). The discrepancy between the values in table 10 for farm No. 161 and those of table 8 is due to the fact that in table 8 only the second series of pasture samples (Design of Experiment) is included, whereas all samples are included in table 10. Average levels of strontium-90 in milk from both groups of cows on farm No. 129 are lower than in milk from the comparable groups on farm No. 161, or in milk from the control farms. This cannot be explained entirely in terms of fertilization and management of the land and may be due to other variables not considered in this experiment.

These studies also confirmed the experimental observations on farm No. 161 with respect to iodine-131. On farm No. 129, levels of iodine-131 in milk from the cows grazing the unfertilized pasture averaged 40 percent higher than in milk from the cows grazing the fertilized control pasture. A limited number of observations were made during September and October of levels of radioiodine in milk from the 10 control farms, which showed good agreement with findings obtained on the experimental farm No. 161 during the same period. An average content of 226 pc per liter of iodine-131 was found in milk from the five control farms rated Poor on fertilization and land management programs, as compared with an average content of 197 pc per liter in milk from the cows grazing unfertilized pasture on farm No. 161 on the same days. Milk from the five farms with Good programs of fertilization and

Table 10. Comparison of the Average Iodine-131 and Strontium-90 Content of Forage

(Concentrations in pc/kg dry wt)

	Farm No. 129 pastures		Farm No. 161 pastures	
	Fertilized	Unfertilized	Fertilized	Unfertilized
I^{131}	900	1,260	820	1,440
Sr^{90}	1,060	1,870	1,020	1,970

land management contained 71 pc per liter, or about one-third as much as was found in the marginally managed farms, and compares with an average content of 48 pc per liter in milk from the cows grazing the highly fertilized experimental pasture on farm No. 161 on the same days.

Examination of Fluctuating Farms; Relationship to Raw Milk Surveillance Sample

In the survey phase of this study, a relatively large group of farms which fluctuated in category from one survey period to the next was not considered because of inconsistent findings. However, it is important to consider this group in the light of subsequent experimental observations. These farms comprised the major part of the 1,000-cow St. Louis milkshed, which had been consistently higher in strontium-90 than the other eight similar milksheds in other areas (4, 5). An understanding of this group helped to dispel the "mystery" concerning the St. Louis raw milk sample.

A phenomenon not anticipated in the design of the surveys became apparent in the findings of the 3-week study of farms No. 161 and 164 in the fall of 1961, and was confirmed by the experimental studies of 1962. This represented a rather wide random daily fluctuation of levels of strontium-90 in milk from any group of cows, which can best be seen in figure 3, where individual daily levels are indicated, and is also evident in figure 4 (even though in the latter case daily fluctuations are leveled somewhat because weekly averages are shown). Since each survey period required approximately 3 weeks to visit all 66 farms, and since the milk sample from each farm was taken on the day the farm was visited, it is likely that this random daily fluctuation in the samples accounts in part for the variation in category from one survey period to the next. Average values of all five surveys would therefore appear to provide a more accurate representation of the categories of these farms than individual values. Table 11 shows the levels of strontium-90 in milk from these farms at each survey with the average for the five survey periods. Some farms went out of the dairy business during the study or became otherwise unavailable and therefore were not included. Inspection of the data shows that nearly all of these farms would fall into the group II, or Intermediate, category of table 3, when averages of all five surveys are considered.

Table 11. Strontium-90 in Milk from Farms Changing Category Between Survey Periods

(Concentrations in pc/l)

Farm No.	Survey Period					Average
	March 1960	August 1960	November 1960	March 1961	June 1961	
101	15.9	13.6	12.0	12.2	8.7	12.5
123	27.4	21.2	14.9	15.2	9.9	17.7
136	42.6	18.2	14.2	18.7	8.7	20.5
187	15.9	19.4	8.3	13.8	16.9	14.9
199	12.2	18.1	12.2	19.7	14.8	15.4
312	17.7	9.7	13.1	9.1	12.7	12.5
315	37.0	18.7	27.9	7.1	16.7	21.5
450	21.2	16.9	12.4	5.1	7.6	12.6
564	14.0	23.5	17.5	7.0	17.2	15.8
585	21.8	13.3	10.8	14.6	16.5	15.4
954	17.8	14.8	14.0	15.7	14.1	15.3
960	6.3	26.8	9.9	15.4	11.0	13.9
999	39.7	16.4	12.0	6.6	15.2	18.0
121	25.8	16.6	18.6	17.9	13.2	18.4
124	13.0	13.2	17.8	8.4	10.6	12.6
160	35.5	20.0	16.2	13.2	12.7	19.5
162	29.5	16.8	8.8	8.8	18.4	16.5
168	22.6	11.2	11.6	15.8	14.9	15.2
163	14.9	16.6	12.7	9.4	11.3	13.0
165	19.8	15.9	20.9	20.1	21.9	19.7
169	23.9	17.0	12.0	8.0	14.6	15.1
373	24.4	10.0	4.2	7.3	10.6	11.3
374	33.0	20.7	19.1	12.8	14.7	20.1
387	12.4	13.2	13.1	9.2	16.7	12.9
389	7.5	19.8	15.4	14.8	22.9	16.1
391	22.0	14.5	8.7	6.9	15.2	13.5
392	23.6	17.6	13.6	13.2	10.6	15.7
393	17.5	17.3	11.8	7.3	13.8	13.5
406	11.4	17.3	9.2	7.2	22.7	13.6
404	20.7	20.3	15.5	13.9	20.7	18.2
408	17.5	8.9	7.5	8.3	11.1	10.7
731	29.1	32.8	16.9	17.2	11.5	21.5
740	12.1	20.4	12.6	12.6	12.4	14.0
Average	21.4	17.3	13.5	11.9	14.3	15.7

A review of the survey data pertaining to farming practices on these farms, viewed in the light of the experimental results, furnished more insight into these farms. The group I and group III farms (table 3 and figure 1), and the conditions set up in the experimental phase of the study, represent extreme situations of

either Very Good or Very Poor with respect to land management and fertilization. Many of the fluctuating farms, however, had mixed situations which apparently represented programs of improvement in progress but not yet completed. Some of these farms, for example, had Very Good management programs with respect to hay, Fair with respect to silage crops, but had not yet had the time nor financial resources to begin improvements of pasture. Various other combinations of Good and Poor management were observed, which might be averaged as Fair for any individual farm. These data are summarized in table 12. It can readily be seen how cows on a given farm could, for example, consume pasture containing a relatively high strontium-90 content because of inadequate fertilization during the summer, and during the winter months

Table 12. Number of Farms Using Different Levels of Fertilization on Three Basic Crops from a Group of 33 Farms

(Amounts of fertilizer in lbs/acre)

PASTURE

Rarely or never	Minimally every 3 to 5 years	200 lbs. or more of 12-12-12 when planted; top dressed at least every 2 years	According to soil analysis when planted; top dressed at least every 2 years
13	13	5	2

HAY

Purchased, no information	Rarely or never	Minimally every 3 to 5 years	200 lbs. or more of 12-12-12 when planted; top dressed at least every 2 years	According to soil analysis when planted; top dressed every 2 years
8	2	8	7	6

SILAGE
(Corn or sorghum)

150 lbs. of 12-12-12 or less when planted	150 to 200 lbs. of 12-12-12 when planted	Over 200 lbs. of 12-12-12 when planted	According to soil analysis when planted
5	11	2	2

consume hay from a new, well-fertilized field with a relatively low strontium-90 content. If, in addition, such cows take some pasture on warm days during the winter months, an additional increment of the radionuclide which cannot be assessed quantitatively may be ingested.

The daily fluctuations discussed above, and the observation of mixed quality of management on this group of farms are sufficient to explain the fluctuations in category from one survey period to the next, which characterized the majority of farms in the 1,000-cow St. Louis milkshed.

The experimental demonstration that land management and fertilization influence the concentration of radiostrontium in milk offers a possible explanation for the consistently high levels of strontium-90 in the St. Louis raw milk sample as compared with the raw milk samples from the other eight 1,000-cow milksheds and with the St. Louis processed milk sample (4, 5). Comparison of the data of table 12 with the criteria of table 1 shows that the average fertilization and land management practices in the 1,000-cow St. Louis milkshed fell short of optimal. The consistent difference between levels of strontium-90 in the raw milk sample and processed milk sample was related to the fact that 60 percent or more of the milk produced came from 30 percent or less of the farms. The higher radionuclide levels in milk from the more numerous marginal dairy farms were diluted in processing with the larger quantity of milk from the smaller number of well-fertilized, high-producing dairy farms. Although the possibility that a unique weather pattern may have contributed to this situation has not been ruled out, it does not appear likely that this was a major cause of the consistently high levels of strontium-90 in the St. Louis raw milk sample.

DISCUSSION

It has been shown experimentally that levels of iodine-131 and strontium-90 in milk can be effectively moderated by good management and optimal fertilization of the land which provide a luxuriant, rapidly growing pasture crop throughout the grazing season. The mechanism involved was indirect--these radionuclides were diluted by the larger mass of the more rapidly growing, well-fertilized plants. (It may be speculated that methods of obtaining increased forage yield per unit area of land by methods other than by fertilization

would be equally effective. Such possibilities should be verified experimentally.) The relationship between levels of these radionuclides in milk and levels in pasture plants is in agreement with the findings of a survey of four farms near Brainerd, Minnesota (13) where farm-to-farm differences in the strontium-90 content of milk were linearly correlated at a highly significant level with farm-to-farm differences in hay. In view of some data suggesting that production of fodder was greater on the two "low" farms in the Brainerd study than on one of the "high" farms, it is possible to speculate that the farm-to-farm differences in the strontium-90 content of the hay on those farms may have been accounted for partly in terms of more rapid growth of fodder on the "low" farms. However, other mechanisms involving topography of the land may also have influenced the situation.

Concentrations of radionuclides in plants growing on fertilized land were found to be lower than in plants growing on unfertilized land. This agrees with an earlier report by Libby (14) and more recent reports by Menzel (15), Guliakin and Yudintseva (16), and Guliakin and Korovkina (17) that addition of mineral and/or organic amendments to the soil can favorably modify the uptake of fission products by growing plants. The data, however, do not indicate whether similar or different mechanisms are involved. It is important to point out that efforts to moderate the uptake of fission products by plants through the use of fertilizers should be based on soil analysis, and all requirements for good growth should be supplied in a balanced mixture.

The close agreement between results from experimental farm No. 161 and findings from the group of 10 nonmanipulated control farms shows that the results of the experimental manipulations are related in a very real way to similar situations outside the immediate experimental circumstances, and suggests that the fission product content of the milk from the 5 "high" farms could be similarly reduced by application of the same principle. The implications are of utmost importance since they open the way for the individual dairy farmer to exercise some degree of control over the levels of fission-product radioactivity in the milk produced, using only the materials and methods which are routinely at his disposal. All of the farm work of this study was performed by the dairy farmers who owned or operated the experimental farms, which were not greatly different from many other dairy farms in the same area. All farm materials used were purchased through a

source normally available to any farmer--the Missouri Farmers' Association Produce Exchange. The use of a sophisticated research farm was avoided. (This refers to such farms as are found at land-grant colleges, to sophisticated materials not ordinarily available to the average farmer, or to sophisticated farming techniques not well understood and perhaps not trusted by the average farmer.) The point is made that it is within the capacity of any dairy farmer to utilize the experimental conditions described in this study.

During the past decade, dairymen and milk consumers alike have been subjected to conflicting statements, attributed to the scientific community, pertaining to the deposition and possible effects of fission products from nuclear weapons tests. The consumer, apprehensive of the possible effects of any increase in radiation to which he might be exposed, and confused by the conflicting reports which he hears, has reduced the amount of milk he consumes. The dairyman, in turn, has watched helplessly as his market dwindled (18). Obviously, this situation is not in the best interests of either the milk consumer or the dairyman. Particularly in the case of growing children, and to a lesser degree for the population as a whole, there is no adequate nutritional substitute for milk. If the dairy farmer can effectively reduce the levels of fission products in milk (without resorting to such procedures as ion exchange), the apprehension of the milk consumer will gradually abate, the consumption of milk will increase, and the milk market will tend to return to normal. These considerations are important, both in terms of public health and in terms of economic health. In terms of public health, the responsibility for controlling limits of public exposure to radiation must be met by public health agencies. This control must be accomplished, if possible, without disrupting the nutrition and dietary habits of the population or disrupting the economy of a region or industry. From the point of view of economy, the use of fertilizers in the amounts indicated by this study is costly. However, the ultimate consequence of such fertilization is an increase in milk production to the capacity of the cows, which is a rare situation on the marginal dairy farm. Receipts from this increase in milk production would more than offset the costs of the fertilizers and seeds in the course of a year, if a normal market for milk prevails. Good growth of appropriate forage plants, resulting in luxuriant crops, is not only a key to the moderation of levels of fission products in milk; it is also a key to the profitable operation of a dairy farm.

These studies could be a foundation for future cooperative efforts between public health agencies and the dairy industry to exercise a degree of control over fission-product contamination in milk. Just as cooperative efforts in the past have led to remarkable improvements in the sanitary qualities of milk, it can be expected that improvements can be effected in control of radiological contamination of milk, if atmospheric fission-product contamination makes such efforts mandatory. Application of these methods will not affect the taste or quality of milk, should not affect its cost, and in the long run should be actually beneficial to the economy of the dairying region involved.

SUMMARY AND CONCLUSIONS

A series of surveys of the farms which made up the 1,000-cow milkshed of the St. Louis raw milk surveillance sample indicated that levels of strontium-90 in milk are related inversely to the quality of fertilization and land management in a mechanism involving the growing of feeds. Similar findings were obtained in the case of iodine-131 in milk. Subsequent experimental modification of the fertilization and land management practices on a marginal dairy farm demonstrated that the levels of iodine-131 and strontium-90 in milk from atmospherically transported nuclear test debris could be reduced by a factor of 50 percent or more throughout the entire pasturing season by grazing cows on abundant well-fertilized pastures. A causal relationship between optimal fertilization of the land and decreased radionuclide content of the milk was established. Forage from the well-fertilized land contained lower concentrations of these radionuclides than did forage from unfertilized land. A mechanism of dilution, in which concentrations of deposited fission products in pasture were inversely proportional to the dry weight of plant material per unit area of land, was shown to account for the findings in milk. Various other possible mechanisms which might account for the experimental results, including variance in rainfall, dilution in diet by ground grain mixtures, dilution in larger volumes of milk, and variance in discrimination on a strontium-to-calcium basis were ruled out. The data suggest that the consistently high levels of strontium-90 in the St. Louis raw milk sample (as compared with the other eight 1,000-cow milksheds and with the St. Louis processed milk sample) were not the result of an unusual weather pattern, but were due to less than optimal

fertilization practices leading to ingestion by cows of the fission products taken up over greater areas of land than in other situations where heavy fertilization was more routine. The principle of optimal fertilization and management of the land provides a basis for possible future cooperative efforts between public health agencies and the dairy industry toward control of limits of fission-product contamination in milk, should such control measures be necessitated by substantial or sustained releases of fission products into the uncontrolled environment.

ADDENDUM

In order to establish whether or not these findings could be utilized outside the dairying region of Missouri, a confirmatory experiment was conducted in southern Mississippi, within the New Orleans milkshed. Consistently high levels of strontium-90 were found in the New Orleans processed milk sample during 1962 and early 1963 (19). Climatic conditions, soil, and crops are substantially different in this area than in Missouri.

The design of this experiment was basically the same as that conducted in Missouri. On a marginally managed farm, where most of the land had not been fertilized within 5 years, the pasture was divided into experimental and control sections. Experimental pastures were fertilized according to soil analysis, with an additional 10 percent increment as a "booster." Control pastures were untreated. Forage crops on the control pasture included: Bahia grass, Bermuda grass, sweet clover, and some crimson clover. Forage crops on the experimental pasture included: Rye grass, Bahia grass, crimson clover, Kobe lespedeza, and fast-growing drought-resistant hybrid Lindsay 77 ("Sudex"). The cows on experimental and control pastures were switched once. Milk samples were collected twice each week (Tuesday and Thursday), from April 18 through August 15, 1963. Preliminary findings from this experiment (which will subsequently be reported in greater detail) are presented in figure 5. The group A cows were on experimental pasture and the group B cows on control pasture during this period. The findings obtained in the St. Louis milkshed are confirmed in the New Orleans milkshed. It is of interest that the differences in levels of strontium-90 in milk from the two groups of cows narrowed markedly during May, when a spring drought caused the experimental pastures to deteriorate, then widened with the resumption of rainfall.

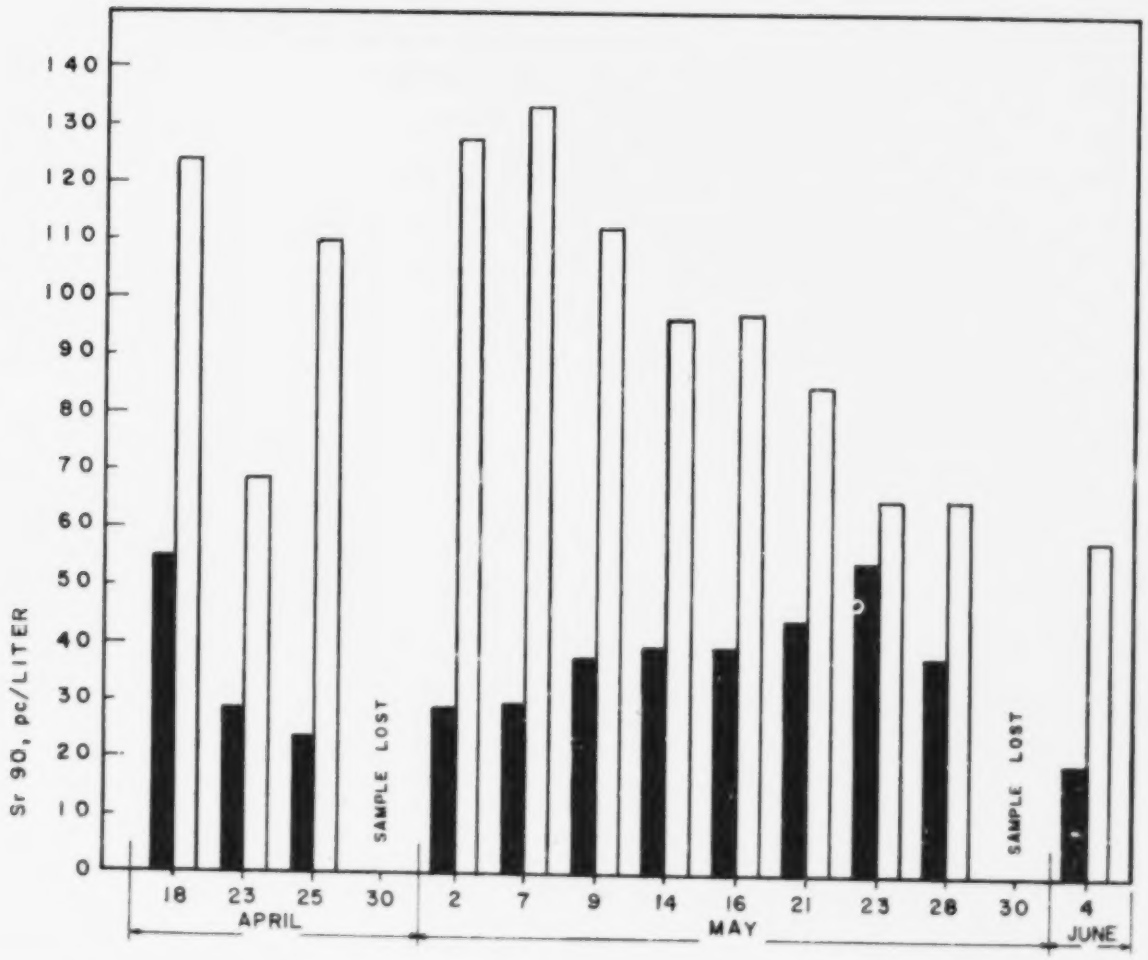

Figure 5. Levels of strontium-90 in milk from group A cows (solid bars) and from group B cows (open bars) on farm No. 103

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APPENDIX I

Radiochemical Determination of Strontium-90

A.$_1$ PREPARATION OF FEED SAMPLES

1. Wet weight of sample is determined. Sample is dried overnight or longer at 110^o in convection oven. Dry weight is determined.

2. Sample is quantitatively transferred to a 5-gallon galvanized bucket. Approximately 500 ml of fuel oil are added and the sample is ignited out of doors over a bed of glowing charcoal.

3. The burned sample is transferred to a 350 ml Vycor dish and ashed in the muffle furnace at 450^o for 2 hours. Heating is continued at 650^o for 3 hours. The ash weight is recorded.

B.$_1$ PURIFICATION OF STRONTIUM AND BARIUM

4. Quantitatively (with aid of a little 6N HCl) transfer 10.0 gm of ash to centrifuge cup. Add 1 ml 2N strontium carrier and 1 ml 0.2N barium carrier.

5. Dissolve ash in a minimum quantity of 6N HCl with heating (almost to boiling).

6. Centrifuge, decant supernatant and save.

7. Repeat step 5.

8. Centrifuge. Decant supernatant and pool with supernatant of step 6. Discard residue.

9. Add 100 ml of saturated oxalic acid. Stir.

10. Adjust the pH to 3 with the careful addition of conc. NH_4OH. Allow to stand for a few minutes. Go to step 11 below.

A.$_2$ PREPARATION OF LIQUID MILK SAMPLES

1. Shake sample well. Measure out 1.0 liter into a 3-liter beaker. Add 1 ml strontium carrier and 1 ml barium carrier. Stir and allow to stand for a few minutes.

A.$_2$ PREPARATION OF LIQUID MILK SAMPLES (continued)

2. With stirring add 1 liter of 24 percent trichloroacetic acid. Allow to stand for 30 minutes.

3. Filter through a fluted filter paper (Whatman No. 1). Discard protein residue.

B.$_2$ PURIFICATION OF STRONTIUM AND BARIUM

4. Add 100 ml of saturated oxalic acid. Stir.

5. Adjust the pH to 3 with the careful addition of concentrated NH_4OH. Stir. Allow to stand for at least 24 hours to permit the alkaline earth oxalates to settle.

6. Siphon off the bulk of the supernatant. Quantitatively transfer the remainder of the mixture to a 250 ml centrifuge cup.

7. Go to step 11 below.

11. Centrifuge at 1800 rpm for 10 minutes. Discard supernatant.

12. Wash twice with demineralized water. Suspend and centrifuge.

13. Transfer as nearly quantitatively as practicable, to a 100 ml Vycor dish or Ni crucible.

14. Heat in convection oven at 110° until dry.

15. Transfer to muffle furnace. Heat at 450° C. for 2 hours.

16. Continue heating at 650° for 3 hours. Allow to cool.

17. Transfer quantitatively with a small volume of 1N nitric acid to a 250 ml beaker (previously calibrated to 30 ml).

18. Evaporate to dryness.

19. Add 10 ml of conc. HNO_3; evaporate to dryness.

20. Add 20 ml of conc. HNO_3; evaporate to dryness.

21. Add 50 ml of conc. HNO_3; evaporate to 30 ml.

22. Transfer to 40 ml centrifuge tube. Cool in an ice bath for 30 minutes.

B.$_2$ PURIFICATION OF STRONTIUM AND BARIUM (continued)

23. Centrifuge at 3000 rpm for 10 minutes. Discard supernatant liquid.

24. Dissolve in 2 ml of demineralized water. Warm to promote solution. Insoluble silicates may be present at this point. If present, centrifuge at 3000 rpm. Transfer supernatant to a new centrifuge tube.

25. Add 10 ml fuming HNO_3 and continue warming for another 10 minutes.

26. Cool in an ice bath for 30 minutes.

27. Centrifuge at 3000 rpm for 10 minutes. Discard supernatant liquid.

28. Dissolve in 20 ml of water. (If solution is not complete add a few drops of HCl.)

29. Make alkaline with 6N NH_4OH and reppt. strontium and barium with excess 3N Na_2CO_3.

C. SEPARATION OF BARIUM

30. Dissolve the ppt. in about 2 ml of 6N HCl. Warm to promote solution and to remove any CO_2.

31. Neutralize the solution with 6N NH_4OH, using methyl orange indicator.

32. Dilute to 20-25 ml with demineralized water. Add 10 ml ammonium acetate buffer.

33. Place the solution in a hot water bath. Add 1 ml of 1N Na_2CrO_4 and digest in a hot water bath for 5 minutes. Add 1-2 drops of 1N $BaCl_2$ and continue digesting for another minute or two with occasional stirring.

34. Centrifuge at 3000 rpm for 10 minutes. Decant supernatant and save for radiostrontium determination.

35. Add 5 ml demineralized water to $BaCrO_4$ ppt., warm for 1-2 minutes, cool, centrifuge at 3000 rpm for 10 minutes and discard washings.

C. SEPARATION OF BARIUM (continued)

36. Filter through a Millipore filter apparatus using a filter paper which was previously weighed with a planchet.

37. Determine beta activity in low level beta (anticoincidence) counter. Alpha activity may be determined in the internal proportional counter as a rough index of radium-226 content.

D. SCAVENGING OF RARE EARTHS

38. Add 6N NH_4OH to supernatant of step 34 to make solution alkaline and ppt. strontium with excess 3N Na_2CO_3.

39. Centrifuge and discard supernatant. Wash ppt. with water containing a drop or so of 3N Na_2CO_3 and discard washings.

40. Dissolve $SrCO_3$ ppt. in 3 ml of 6N HCl.

41. Add 20 ml of water, 1 ml of mixed rare earth carrier. Heat and make alkaline with 6N NH_4OH.

42. Filter through No. 1 Whatman paper and discard ppt.

43. Add 1 ml of mixed rare earth carrier and allow to digest 5-10 minutes. Record time of pptn. (this begins time of ingrowth of yttrium-90).

44. Filter through No. 1 Whatman paper and discard ppt.

45. Add sufficient 3N Na_2CO_3 to filtrate for complete pptn. of strontium carbonate. Centrifuge. Decant and discard supernatant.

46. Store ppt. for at least 2 weeks (in that time 97 percent of secular equilibrium is attained).

E. EXTRACTION OF YTTRIUM-90

47. Dissolve ppt. in 2 ml 4N HNO_3.

48. Cool in an ice bath and add with stirring 10 ml of fuming nitric acid from an automatic pipet.

49. Centrifuge and transfer supernatant into a 60 ml separatory funnel. Retain ppt. for determination of total radio-strontium and chemical yield (see step 57).

E. EXTRACTION OF YTTRIUM-90 (continued)

50. Add 3 ml 4N HNO_3 to separatory funnel to bring aqueous phase to 15 ml.

51. Add 10 ml equilibrated TBP (tributyl phosphate) and shake thoroughly for 10 minutes. (This starts time of decay of yttrium-90.) Note time of addition.

52. Wash organic phase once with a 5 ml portion of 14N HNO_3. Discard washing.

53. Back-extract organic phase 3 times (5 minutes each time); twice with 5 ml portions of demineralized water; once with 5 ml of 0.1N HNO_3.

54. Transfer aqueous phase directly to stainless steel planchet in a stepwise manner and evaporate to dryness on a warm hot plate.

55. Eliminate final trace of TBP by direct flaming of the planchet over a Meeker burner.

56. Determine activity in a low background anticoincidence beta counter.

F. DETERMINATION OF CHEMICAL YIELD OF STRONTIUM CARRIER

57. Dissolve ppt. from step 49 in demineralized water.

58. Transfer to a 25 ml volumetric flask. Make up to volume and mix.

59. Transfer 10 ml of this solution to a 100 ml volumetric flask for the determination of chemical yield. Make up to volume and mix.

60. Standard solutions, containing 1.00 to 10.0 meq/liter of Sr^{++} are poured into 5 ml Beckman beakers.

61. A portion of the unknown solution of step 58 is poured into a 5 ml Beckman beaker.

F. DETERMINATION OF CHEMICAL YIELD OF STRONTIUM CARRIER (continued)

62. Set wavelength at 460 mu. Fine adjustment for maximum response should be made periodically. Instrument should be operated at the 0.1 setting.

63. Turn on the flame. Oxygen and hydrogen pressures for operation vary from one instrument to another.

64. Prepare a standard curve, using the 1.00-10.0 meq/liter standards for each batch of analyses, adjusting the slit width and sensitivity so that the 10.0 meq/liter standard reads 100% T. Checks against instrument drift are made by returning to the 10.0 meq/liter standard after reading several samples. This should read within the limits 98-102% T.

65. Read each unknown. Instrument must be checked against standards periodically and corrected for drift.

66. Convert instrument readings to meq strontium recovered, using the standard curve.

G. CALCULATIONS

67a Strontium-90 in milk

$$\text{pc } Sr^{90}/l = \frac{\text{net cpm}}{2.22\ E\ I\ R\ V}$$

E = efficiency of counting yttrium-90
I = ingrowth factor for yttrium-90
R = recovery as determined by flame photometry
V = volume of sample

67b Strontium-90 in feeds

$$\text{pc } Sr^{90}/kg = \frac{\text{net cpm } (WA/WS \times 1000/W)}{E\ I\ R\ 2.22}$$

WA = ash weight
WS = sample weight
W = dry weight
E = counter efficiency
I = ingrowth factor for yttrium-90
R = recovery as determined by flame photometry

APPENDIX II

Determination of Iodine-131 in Liquid Milk

1. Measure out 3.5 liters of milk into the polyethylene Marinelli beaker.

2. Turn Channel Group knob on gamma spectrometer to the last position to the left and Readout knob to "CRT." Then turn Control knob to "readout" and push the button marked "erase."

3. Place cesium-137 standard on the detector. Turn Control knob to "analyze" for 5 to 10 seconds.

4. Turn Control knob to "readout." Check oscilloscope. The highest point in the cesium-137 peak should be at channel 33. The point to the right of the highest point should be between the highest point and the point to the left. Adjust Fine Gain "A" and reanalyze until this configuration occurs.

5. With Control knob on "readout" push "erase" button to clear memory.

6. Place container of sample over detector.

7. With Time knob at "100" turn control knob to "analyze." Set timeclock to ring after 52 minutes.

8. Fill out type-out sheet on IBM typewriter.

9. After 50 minutes or so, the light over "readout" on the Control knob should be on. Move the Control knob so that the light at the "stop" position is on. Turn Channel Group knob to "0-256" and Readout knob to "type."

10. Turn Control knob to "readout." Data will type out.

CALCULATIONS:

1. Sum channel groups 15-20, 22-28, 30-36, and 70-76, respectively.

2. Divide each sum by the amount of time sample was analyzed (50 minutes).

3. Subtract weekly background averages from the corresponding channel groups.

APPENDIX II (continued)

4. Sum 15-20 minus background equals N_I, sum 22-28 minus background equals N_L.

5. Sum 30-36 minus background equals N_C, sum 70-76 minus background equals N_K.

6. Substitute these values into the formula:

$$I = 19.9\ N_I - 14.4N_L - 3.73\ N_C - 2.68\ N_K$$

7. Divide result by 7.77 and correct for decay of iodine-131 since collection of sample. Result is in pc/liter at time of sample collection.

APPENDIX III

Determination of Calcium in Liquid Milk

1. Place 50.0 ml of milk into a 250 ml beaker. Add 50.0 ml 24 percent trichloroacetic acid. Stir.

2. Filter through No. 1 Whatman paper into any suitable container.

3. Transfer two 10.0 ml aliquots into two 40 ml centrifuge tubes.

4. Add 10 ml of saturated oxalic acid. Mix.

5. Adjust the pH to 3 with 6N NH_4OH using methyl violet indicator.

6. Mix and centrifuge at 3000 RPM for 5 minutes. Discard supernatant. Wash twice with demineralized water.

7. Dissolve ppt. in a minimum volume of 2.5N HCl with warming. Transfer quantitatively to a 125 ml Erlenmeyer flask. Bring volume to about 25 ml with demineralized water.

8. Heat nearly to boiling. Titrate against 0.050N $KMnO_4$. The first permanent pink color defines the end point. Similar titrations must be performed against a blank solution containing only HCl and water, and against appropriate calcium standards with each batch of unknowns.

   The volume of the blank titration must be subtracted from all other titrations.

APPENDIX III (continued)

CALCULATIONS:

V = volume of standard $KMnO_4$, corrected for blank titration.

$$V \text{ ml} \times \frac{0.050\text{meq}}{\text{ml}} \times \frac{2}{1} \times \frac{1{,}000 \text{ ml/liter}}{20 \text{ ml}} \times \frac{20.04\text{m}}{\text{meq}} \times \frac{1\text{g}}{1000 \text{ mg}} = \text{g/liter}$$

Or, V ml x 0.050 x 2.004 = g/liter

APPENDIX IV

Determination of Calcium in Feeds

1. Weigh 0.500g ashed sample in a centrifuge tube.
2. Add approximately 5 ml of 6N HCl to sample. Heat in water bath. Decant into another centrifuge tube.
3. Repeat step 2 twice, decanting into same centrifuge tube.
4. Add 15 ml saturated oxalic acid. Mix well.
5. Go to step 5 of procedure for calcium in milk.

CALCULATIONS:

V = volume of standard $KMnO_4$, corrected for blank titration.

$$V \text{ ml} \times \frac{0.05\text{meq}}{\text{ml}} \times F \times \frac{20\text{mg}}{\text{meq}} \times \frac{1\text{g}}{1000\text{mg}} = \text{g/kg}$$

$$\frac{V \times F}{1000} = \text{g/kg}$$

where:

$$F = \frac{WA}{WAS} \times \frac{1000}{W}$$

W = dry wt
WA = ash wt
WAS = sample wt

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